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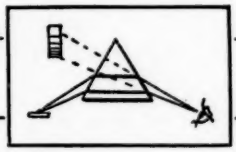

OCTOBER
1934

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Journal of the
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VOLUME 24 OCTOBER, 1934 NUMBER 10

The Optical Constants of Polished and Sputtered Molybdenum Surfaces

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(Received May 22, 1934)

The optical constants of molybdenum are determined by Drude's method for opaque films sputtered in air and in hydrogen, and for mirrors of massive metal, at wavelengths 0.578μ, 0.546μ and 0.436μ. The indices of refraction and absorption, and the computed reflectivity are found to vary with the conditions of sputtering, and to be considerably less than for the massive metal. To eliminate any possible effect due to surface films formed by chemical action of the atmosphere, the final stage of polishing one of the bulk metal mirrors is carried out under kerosene, and the mirror is observed while immersed in kerosene. The results are not appreciably different when the same mirror is observed in air.

PLANE polarized light reflected from a metallic mirror becomes, in general, elliptically polarized. By observation of this elliptically polarized light, two characteristic constants of the mirror, the index of refraction n and the index of absorption κ may be determined. This method was applied to mirrors of polished and of sputtered molybdenum.

At first a mercury "Lab-Arc" was used as source. For greater intensity, this was later replaced by an air-cooled capillary mercury arc.[1] After passage through a constant deviation monochromator, the light was plane polarized by a Nicol, set successively in the four positions for which the plane of polarization made an angle of 45° with the plane of incidence. The light was then reflected by the molybdenum surface. A Babinet-Soleil compensator restored the resulting elliptically polarized light to plane polarized light whose azimuth was determined by an analyzing Nicol equipped with a Zehnder half-shade.[2] Each set of observations depended on 64 matches of the half-shade field.

The quantities observed were the angle of incidence, the phase change introduced by the compensator, and the azimuth of restored plane polarization. Substitution of these values into Drude's formulas[3] yielded n, κ and the reflectivity R. In this system of notation the complex refractive index is $n[1-(-1)^{\frac{1}{2}}\kappa]$.

PREPARATION OF MIRRORS

(a) Sputtered mirrors

The sputtering chamber was a one gallon jar, arranged as described by Cartwright.[4] The surfaces to receive the sputtered molybdenum rested on a glass table 3 cm below the cathode, and about 50 cm from the aluminum anode. A small 5000 volt "luminous tube" transformer main-

[1] A. Allen and E. Ponder, J. Physiology **66**, 37 (1928).

[2] Zehnder, Ann. d. Physik **26**, 985 (1908).

[3] P. Drude, *Lehrbuch der Optik*, 2nd edition, p. 344.

[4] C. H. Cartwright, R. S. I. **1**, 759 (1930).

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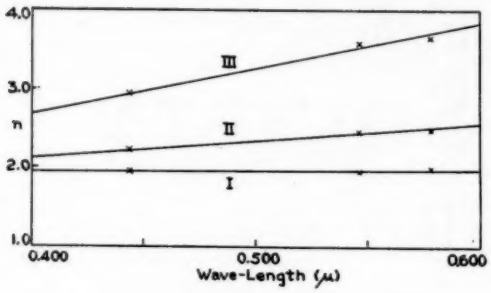

FIG. 1. Index of refraction n plotted against wave-length. Curve I. Molybdenum surface sputtered in hydrogen. Curve II. Molybdenum surface sputtered in air. Curve III. Surface of massive molybdenum.

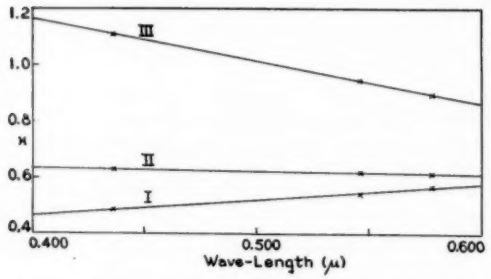

FIG. 2. Index of absorption κ plotted against wave-length. Curve I. Molybdenum surface sputtered in hydrogen. Curve II. Molybdenum surface sputtered in air. Curve III. Surface of massive molybdenum.

tained the gaseous discharge. No rectifier was used.

Three mirrors were sputtered on glass with air as the residual gas. To minimize oxidation the pressure was maintained just above that value at which the discharge ceased to pass. Opaque mirrors were obtained in 13 hours.

Two mirrors were sputtered on glass in electrolytic hydrogen. The pressure in the cathode chamber was regulated so that the Crookes dark space extended about to the mirrors.[5] Opaque mirrors were obtained in 35 hours.

(b) Mirrors of massive metal

Mirrors were prepared from disks of molybdenum, 1/2″ in diameter and 1/4″ thick, stated by the manufacturer to be 99.9 percent pure. These blanks were rubbed by hand on glass

[5] E. O. Hulburt, R. S. I. **5**, 85 (1934).

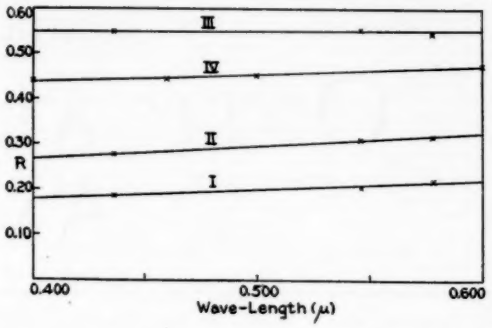

FIG. 3. Reflectivity R plotted against wave-length. Curve I. Molybdenum surface sputtered in hydrogen. Curve II. Molybdenum surface sputtered in air. Curve III. Surface of massive molybdenum. Curve IV. Reflectivity of massive molybdenum by direct measurement. Data from Coblentz.[8]

plates with successively finer grades of carborundum wet with kerosene. They were then buffed on a cotton wheel with occasional application of a paste of optical rouge and water. One mirror was observed without further treatment.

A second mirror was further polished for 2 hours on a rotating lap of beeswax (apiarist's "medium brood foundation" on a plaster base[6]) with rouge and kerosene.

A third mirror, after buffing, was given a brilliant polish by 2 hours' application to a rotating lap of soft cloth, with a thick paste of rouge and water. After a series of optical observations, this mirror was rubbed for 2 hours on a lap of chamois immersed in kerosene, with a very little rouge. It was hoped that this treatment would remove any oxide film which might be present. The mirror was thereafter kept immersed in kerosene. During the observations it was mounted in a kerosene cell with Pyrex windows which were normal, respectively, to the incident and reflected rays. After multiplication of n by the index of refraction of the kerosene, the results agreed within 2 percent for n, and within 1 percent for κ, with the values obtained when the same mirror was observed in air.

The mirrors of massive metal were all found to satisfy Drude's criterion[7] for freedom from contamination: the phase change introduced by

[6] *Amateur Telescope Making*, A. G. Ingalls, Editor, p. 175, Scientific American Pub. Co., 1928.

[7] P. Drude, Ann. d. Physik **39**, 481 (1890).

reflection was as great as for a freshly filed surface.

Results

Table I gives averages computed from 37 sets of observations at angles of incidence between 45° and 70°. These data are plotted in Figs. 1, 2 and 3. Curve IV, Fig. 3, was plotted from data of Coblentz,[8] obtained by direct measurement of the reflectivity of molybdenum.

Table I.

		n	κ	R
Mirrors sputtered in hydrogen				
Wave-length	0.436μ	1.95	0.484	0.187
	0.546μ	1.96	0.542	0.206
	0.578μ	1.99	0.566	0.220
Mirrors sputtered in air				
Wave-length	0.436μ	2.22	0.626	0.278
	0.546μ	2.46	0.619	0.312
	0.578μ	2.48	0.614	0.318
Mirrors of massive metal				
Wave-length	0.436μ	2.95	1.113	0.551
	0.546μ	3.59	0.948	0.556
	0.578μ	3.65	0.897	0.548

[8] W. W. Coblentz, Bur. Standards Bull. **7**, 107 (1911).

The residual gas appears to play an important rôle in the optical constants of the sputtered surfaces. The mirrors sputtered in hydrogen have much lower index of refraction and reflectivity than those mirrors which were sputtered in air. Ingersoll and Hanawalt[9] have shown that, in films of nickel sputtered in hydrogen, the crystal lattice may be distended by occluded gas, with striking effect on the physical properties of the films. It is not unlikely that the optical properties of the sputtered mirrors in this research are altered by this phenomenon. Lauch[10] associates low values of κ with "loose" atomic arrangement, as may be caused by occluded gas. This agrees with the low κ found for sputtered mirrors, and the higher κ for the mirrors of massive metal.

In conclusion the writer desires to express his deep appreciation to Professors H. C. Richards and R. W. Duncan for many helpful suggestions and for their continued interest in this research.

[9] L. R. Ingersoll and J. D. Hanawalt, Phys. Rev. **34**, 973 (1929).

[10] K. Lauch, Ann. d. Physik **74**, 55 (1924).

Complementarism of the Standard O. S. A. and I. C. I. Observers

I. H. GODLOVE, *Washington, D. C.*

(Received July 18, 1934)

I. INTRODUCTION

SINCE complementary color-stimuli form a relatively simple, two-component, system, less complex than the three-component systems generally dealt with in colorimetry, they afford a convenient starting point for the study of some colorimetric relations. The author has published[1] a convenient method for finding the equations describing the wave-lengths of complementary hues. It is of interest to compare with this means the complementarisms of the two observers that have been adopted in this country as standard (the superseded O. S. A. and new I. C. I. observers), and the complementary wave-length data determined by direct experiment.

II. DATA AND EQUATIONS

Since the curves describing complementary wave-lengths are nearly hyperbolae, and it is possible, by a simple change of variables, to plot the data as straight lines,[1] this method of "rectifying the hyperbolae" was used to find the equations. The complementary wave-length pairs were first found by computing the function $(r-r_w)/(g-g_w)$. Here r and g are the coordinates of the spectrum color of the given wave-length on the (r, g)-plot of the tristimulus mixture data, and r_w and g_w are the corresponding coordinates of the "white point." The "white light" to which the complementarism of both the O. S. A. and I. C. I. observers was referred was mean noon sun, since the experimental data of Sinden[2] was referred to this neutral light. The coordinates of this point were taken as $r_w=0.3319$, $g_w=0.3362$, for the O. S. A., and $r_w=0.3372$, $g_w=0.3550$, for the I. C. I. observer. The specification of the neutral light employed for the older data summarized by Helmholtz is somewhat uncertain.

The method for finding the complementary wave-length equations consisted in plotting the values of $\lambda-\lambda_0$ as abscissae and of the ratio $(\lambda-\lambda_0)/(\lambda'-\lambda_0')$ as ordinates, where λ and λ' are the complementary pair of wave-lengths and λ_0 and λ_0' are a particular pair, exactly on the curve, chosen as a reference point. From the slope S and intercept I of the "best" straight line through the plotted points, the values of the constants of the complementary wave-length equations were computed by means of the following relations:[1]

$$(\lambda-a)(\lambda'-b)=c, \tag{1}$$

where

$$a=I/S-\lambda_0, \tag{2}$$

$$b=-(I/S+\lambda_0') \tag{3}$$

and

$$c=-I/S^2. \tag{4}$$

Proceeding in this way, the following equations were found:

O. S. A. $(\lambda-569.74)(494.04-\lambda')=164.46$, (5)

I. C. I. $(\lambda-565.52)(497.78-\lambda')=223.02$, (6)

R. H. Sinden

$$(\lambda-567.62)(497.98-\lambda')=191.70. \tag{7}$$

In the computations, Sinden's red and violet regions of invariant hue were taken as 700 mμ and 400 mμ, respectively. The reference points used for the three sets of data were, respectively: (580, 478.01), (580, 482.41), and (580, 482.5). When the difference between the experimental O. S. A. and I. C. I. complementaries and those computed from Eqs. (5) and (6) were taken, it was found that the average deviation at nine points in each case was less than half a millimicron. These deviations were computed at the following wave-lengths: 660, 640, 620, 600, 580, 480, 460, 440, and 420 mμ.

[1] Godlove, J. O. S. A. **20**, 411 (1930).
[2] Sinden, J. O. S. A. and R. S. I. **7**, 1123 (1923).

TABLE I. *Complementaries of O. S. A. and I. C. I. observers.*

λ	λ′ exp. O. S. A.	λ′ exp. I. C. I.	λ′ calc. O. S. A.	λ′ calc. I. C. I.	λ′ exp. − λ′ calc. O. S. A.	λ′ exp. − λ′ calc. I. C. I.
700		495.54				
680		495.51				
670	492.10	495.47				
660	492.07	495.42	492.22	495.42	+0.15	0.00
650	492.00	495.32				
640	491.83	495.13	491.70	494.79	−0.13	−0.34
630	491.47	494.82				
620	491.05	494.32	490.76	493.34	−0.29	−0.98
610	490.30	493.45				
600	488.91	491.86	488.61	491.35	−0.30	−0.51
580	478.01	482.41	477.97	481.05	−0.04	−1.36
490	607.11	592.97				
480	581.36	577.89	581.45	578.06	+0.09	+0.17
470	576.22	573.04				
460	573.19	571.06	574.57	571.42	+1.38	+0.36
450	571.94	570.00				
440	571.48	569.36	572.78	569.38	+1.30	+0.02
430	571.30	569.00				
420	571.24	568.75	571.96	568.39	+0.72	−0.36
410	571.16	568.72				
400		568.69				
380		568.65				
				Average	0.49	0.46

TABLE II. *Complementaries of Sinden's observers.*

λ	Sinden	λ′ exp. O. S. A.	λ′ exp. I. C. I.	Difference from Sinden's exp. O. S. A.	Difference from Sinden's exp. I. C. I.
		R. H. Sinden			
700	496.5	492.1	495.5	−4.4	−1.0
609	493.5	490.2	493.2	−3.3	−0.3
591	490	486.7	489.3	−3.3	−0.7
586	487.5	484.5	487.0	−3.0	−0.5
580	482.5	478.0	482.4	−4.5	−0.1
480.5	578.5	581.8	578.2	+3.3	−0.3
477.5	576.5	579.7	576.5	+3.2	0.0
474.5	575.5	578.2	575.1	+2.7	−0.4
472	574	577.1	573.9	+3.1	−0.1
466.5	573	574.8	572.0	+1.8	−1.0
459	572	572.9	570.9	+0.9	−1.1
400	570.5	571.1	568.9	+0.6	−1.6
		Sinden's observers, except RHS, AE, and HTW.			
700	494.9	492.1	495.5	−2.8	+0.6
609.5	491.9	490.3	493.3	−1.6	+1.4
581	485.3	479.5	483.4	−5.8	−1.9
400	568.6	571.1	568.9	+2.5	+0.3
			Average[4]	2.9	0.7

III. COMPARISON OF O. S. A. AND I. C. I.

The O. S. A. and I. C. I. curves of actual data are approximately parallel, not crossing each other; in other words, agree at no real point. The difference between them is from 2 to 4 millimicrons. The agreement is somewhat better in the violet than in the red end. The equations give results differing by an average of about 3.2 millimicrons at nine points. The data are given in Table I. In this table, "λ′ exp." means the experimental results converted to refer to the assumed white light.

IV. COMPARISON WITH OTHER EXPERIMENTAL DATA

In Table II is given the experimental data of R. H. Sinden, and of his other observers, excluding AE and HTW, who were found abnormal in some respect.[3] It is seen that the agreement with experiments of Sinden is somewhat better for the I. C. I. than for the O. S. A. data, except that the agreement of the O. S. A. data with Sinden's own (but not with Sinden's other observers) is better at the violet end. Equations for the experimental results summarized by Helmholtz have been given,[1] but they are not reproduced here because uncertainty concerning the neutral light precludes the throwing of much light on the present comparisons.

V. HYPOTHESIS OF MACULAR PIGMENTATION DIFFERENCE

It has been suggested that the difference between the O. S. A. and I. C. I. observers could be explained by a difference in the amount of macular pigmentation of the two, the pigment corresponding to observation through a yellow filter of dominant wave-length about 572 or 574 mμ. Though this seems a very plausible explanation for much of the difference, it cannot be the sole cause, for, although the data from 700 to 590 mμ do agree with the hypothesis that the I. C. I. observer has more macular pigment than the O. S. A., for the region 400 to 470 mμ the reverse is true. Further, the two curves are

[3] With (581, 485.3) as a reference point, the other three points give for the average of Sinden's observers except himself and AE and HTW, the equation:

$$(\lambda - 563.42)(496.29 - \lambda') = 193.21.$$

[4] These average deviations are not as significant as they might have been if the points had been more uniformly distributed through the spectrum.

approximately parallel, instead of crossing, as they should be if the whole difference were macular pigmentation. Sinden's presumably most pigmented observer (HTW) and least pigmented observer (HNE) have curves which differ by about 13 mμ, for the complementary of 660 mμ, and by about 7 mμ, for the complementary of 430 mμ; but the two cross at the point (574.1, 472.4). It is of interest to note that for most of the spectrum the O. S. A. and I. C. I. curves lie well within the (crossing) curves of Sinden's most and least pigmented observers.

VI. Conclusions

The complementary wave-length pairs of the standard O. S. A. and I. C. I. observers have been computed with reference to a certain neutral light, and have been compared with direct experimental data of Sinden having this neutral. It was found that the I. C. I. data agreed with Sinden better than the O. S. A. did. By means of a method of "rectifying the hyperbolae" of the graphs of the data, equations were found describing the data which reproduced it to within less than half a millimicron. The O. S. A. and I. C. I. curves are approximately parallel and differ by 2 to 4 millimicrons from each other; but both in most of the spectrum lie well within the intersecting curves of Sinden's most and least macular-pigmented observers. The data were found not to be consistent with the hypothesis that the difference between the two standard observers is wholly due to greater pigmentation of the I. C. I. observer, though this may explain a large part of the difference.

Colorimetry with Reflection Standards: A Quasi-Psychological Method.—Interconversion of Physical and Psychological Color Specifications

I. H. GODLOVE AND A. E. O. MUNSELL, *Munsell Research Laboratory*

(Received April 26, 1934)

I. INTRODUCTION. METHODS OF COLORIMETRY

IN color work, strongly contrasted with the "physical" methods of visual or photoelectric spectrophotometry (with their advantages of precision, reproducibility, and freedom from the personal equation), is the ideal psychological method, dealing directly with the three attributes of color. The latter states directly "how a color looks to the eye." Intermediate between these are various colorimetric methods, e.g., so-called trichromatic analysis and visual comparison with systems of reflection or transmission standards. The former one, more exactly called the tristimulus method, is a convenient means for translating the results of one method into another. But until recently, an important connecting link has been missing for the complete conversion of one extreme language into another.

We are prone to lose sight of the fact, in view of the well-known advantages of spectrophotometry, that it is a method which measures only, as a function of wave-length, the proportions of energy absorbed, transmitted or reflected by an object under specified conditions; that is, it gives a *stimulus* specification, *not a color* specification. The results of physical measurements are in fact often difficult of interpretation as color. Therefore, the physical data are combined with psychophysical data referring to some standard observer. But even such combined specifications do not tell in visual terms how much a given color is lighter or darker, more vivid or grayer, or redder or bluer, than any other color in hand. They do have a *greater resemblance* than do purely physical measurements to a direct color description. Further, they may be shown to preserve largely the advantages of spectrophotometry and similar methods.

If we exclude from consideration direct description of color in the terms of introspective psychology, as being too indefinite, in the present state of our knowledge, and too lacking in precision to be practicable, all other methods fall into two classes: (1) Spectrophotometry, which gives the identical radiant power actually evoking the color, and thus is free from the personal equation; and (2) all other methods, commonly called colorimetric, which instead of specifying the actual stimulus, specify a synthetic complex stimulus, not in general the same as the actual one, as judged by the condition of "match," thus introducing the personal equation. The latter includes the so-called mono- and trichromatic analyses and the many comparator methods. Examples of the latter are comparisons with the systematic reflection standards of Ridgway, Ostwald, and Maerz and Paul, and the Lovibond transmitting glasses and Arny solutions. A particular set of reflection standards, those constituting the Munsell system, warrants a separate classification, for reasons of convenience which we shall mention.

The quasi-psychological method

From the theoretical point of view, the ideal psychological method has many advantages; but its use involves many practical difficulties. We must set up, for comparison, scales consisting of exemplars of very many colors. These scales must exhibit variations in one of the three attributes of color while maintaining the other two attributes constant. Within any scale, the intervals from color to color must be constant. The gradations must be such as to conform to the visual mechanism of the standard or average normal observer under certain standard conditions. In the ideal case, the number of colors exemplified must be so great that no given color may differ from some one of the standard colors, under the standard conditions, by more than a "least perceptible difference."

There exists no set of standards which may be considered to approximate closely this ideal. In fact, all but one of them depart very widely from it. The Ridgway, the Ostwald, and the Maerz and Paul standards are *systematic* but do not satisfy the fundamental condition that each of the three attributes of color be exhibited on scales in which the other two are maintained constant; in fact, they do not purport to do so. The Munsell system is unique in making an attempt to satisfy this condition. Insofar as the execution of the Munsell charts is perfect in exemplifying what they purport to represent, and insofar as they conform to the properties of the hypothetical average normal observer, they constitute an approximation to the ideal psychological system. They may be considered to present the most practical working scales of reference, and in turn may be referred to more fundamental scales when these are available. It is convenient to think of them, therefore, as constituting a quasi-psychological system, the closest approximation to the ideal which has yet been developed.[1]

In comparing colorimetric methods, questions often arise concerning the accuracy with which a sample may be specified. If the results of one method can be converted into terms of other methods, these questions concern not only precision and reproducibility of results within the one system but also accuracy of the conversion. In the use of the quasi-psychological method provided by the Munsell standards two sources of error must be considered in connection with precision and reproducibility: (1) error of interpolation among the standards forming the system, and (2) errors due to differences between the standards contained in different Munsell books. A third source of error relates to conversion of the results into other terms. It is the purpose of this paper to give results of a check of overall accuracy of the quasi-psychological method covering all three sources of error. Incidental to this check, there is noted the accuracy of determinations made with the Guild colorimeter,[2] and the accuracy of interconversion of results between the quasi-psychological method and another (the "trichromatic") colorimetric method.

II. Method of Procedure

The method of testing the accuracy obtainable by use of the Munsell charts, and incidentally of the contributory data, was to determine hue and chroma for 32 samples in two independent ways. The samples were opaque, nearly matt, painted

[1] The advantages of a quasi-psychological system, calibrated by physical measurements, are obvious after a little thought. Any set of comparator standards has as its chief advantage the direct presentation of exemplars of the colors in question, so that no demands are made on the color memory, which is notoriously bad, or the imagination. With its aid colors may be so specified that we may know at once and unambiguously whether one is lighter or darker, more vivid or grayer, or redder or bluer, and how much, than another. Further, it is only by the use of a language of specification of color couched in the terms of such a method that we are able to reach a solution of most problems which are problems of color *per se*. Spectrophotometry is ideal in most problems in which our primary interest is not the color, but the physical properties of an object or material, as those problems of plant control of the chemical composition, where color is used merely as an indicator of quality. On the other hand, where we have such problems as the study of "fading," the harmonious relation of the color of stock, initial lettering, and print in advertising, the control and selection of the color of lacquers, enamels and paints; and in all cases of liason between the links of the chain, customer, manufacturer, superintendent, foreman, worker; the need of a visually descriptive method of specification is imperative. If a realization of the differentiation of these two classes of problems were more generally effected, it would help greatly in the clarification of many industrial problems. On the theoretical side, the advantage of a complete quasi-psychological system is that it is the final link in the complete correlation and inter-translation of the diverse languages of color, from the extreme physical to the extreme psychological. Chiefly on the practical, but also on the theoretical, side, accurate calibration of standards carries over to the method the main advantages of spectrophotometry, precision and reproducibility, to a considerable degree. When judging this augmented precision, a fact must be recalled. The great possible precision of purely physical methods, when applied to color specification, is only too often a *pseudo*-precision. In many cases, even when the spectral reflection curves of two colors differ from each other at all points by only the thickness of the pen mark, the *color* difference is clearly perceptible under the proper conditions; on the other hand, cases may occur where the curves differ from each other by a thumb's width at some points when the colors are actually indistinguishable.

[2] These measurements, briefly described below, will be reported in a subsequent paper.

papers,—samples of the same kind as those making up the Munsell charts but not painted with exactly the same pigments nor exactly matching any of them.

The first method

The first method consisted merely in the direct visual comparison of the 32 colors with the color standards of the Munsell *Book of Color* in the usual way. This was done by five trained observers for the first 23 samples selected, and by three observers for the remaining 9. The 9 samples were added in order to provide a further check of yellowish colors for which agreement was least close. The "white light" used was obtained by means of Macbeth Daylight lamps of approximately 6100°K color temperature. The light struck the samples at about 45°, and the viewing was approximately normal to the surface; about equal areas of sample and standard were used; and both were surrounded by a mid-gray field. Since, in the second method, only the chromaticity of the colors was determined, the lightness or darkness being of no special interest, only the hue and the chroma (saturation) of the colors is reported.

The second method—indirect method

The second method is based on the relation of Munsell hue and chroma to tristimulus specifications. The latter were computed from accurate spectrophotometric measurements. The relation to Munsell terms was evaluated by measurement of a large number of samples on the Guild colorimeter, followed by appropriate transformations of the data in a way to be reported in detail.[2] The second method therefore consisted of three steps: (1) spectrophotometry, (2) transformation by computation to tristimulus specifications, and (3) transformation to hue and chroma according to the above relation.

The spectrophotometric determinations formed a part of the measurements made a number of years ago in collaboration with the National Bureau of Standards on samples constituting the forerunner of the present Munsell *Book of Color*.[3]

From the spectrophotometric results for each of the 32 samples were computed trilinear coordinates (r, g, b) according to the O. S. A. excitations with O. S. A. average noon sun as illuminant and basic stimulus. The colors thus specified are the ones that would be evoked in the O. S. A. observer by reflection of average noon sun from the samples. Apparent reflectance for average noon sun was also computed. The samples measured spectrophotometrically were not the identical ones used in the first method, but were pieces of paper cut from the same sheets and carefully compared.

The third step was essentially one of interpolation on one, or between two, of nine large graphs, one for each "value" level, showing loci of hue and chroma on the (r, g)-plot, as determined by measurements on all of the colors of the Munsell system with the Guild colorimeter. Value was found by the known relation between value and apparent reflectance. The procedure in the use of the Guild colorimeter was such that differences between observers were very largely, but not wholly, eliminated. Only a general statement of the procedure will be given here. With the colorimeter, additive trichromatic measurements were obtained on about 400 colors by three observers. Measurements were obtained directly in terms of three actual (light and filter) primary lights, roughly red, green and blue. In order to use these measurements for another purpose, the results were converted by computation to terms of another colorimetric system, going through two steps. The first step expressed them in terms of the British (N. P. L.) primary lights (because the filters had been measured in terms of these primaries by the instrument maker); and the second converted the results to expression in terms of O. S. A. excitations, but with Abbot-Priest artificial sun as illuminant and basic stimulus. The final results were plotted on the tristimulus mixture diagram, with the value of r as abscissa and of g as ordinate. These graphs were used for the *interconversion of tristimulus specification and hue and chroma.* It may be noted that the conversion of the present particular problem is only an example of a general

[3] Unpublished material of the Munsell Research Laboratory and the National Bureau of Standards (N. B. S. Test 46,045). The measurements were conducted under the direction of Dr. K. S. Gibson and the use of the results for the present check was suggested by Dr. D. B. Judd.

method (with also the converse conversion) which *constitutes an important new link in the chain connecting a wholly physical with a wholly psychological method of specification of color or its stimulus.*

III. Sources of Error

Several small errors are known to be present in this work. For example, the samples measured spectrophotometrically, colorimetrically in the Guild instrument, and by direct comparison were not identical samples (because of the danger of wear), though cut from the same sheet and compared by the eye of one observer and found to match. The three "white lights" used or assumed in the computations were close colorimetrically but not identical.[4] The same observers were not used throughout the various steps, though some were common to all observations: also it is not known how closely the average visual systems of our observers approximate to the standard visual system assumed in reducing the spectrophotometric results. The Guild measurements were not completely free from personal variations, and there were also important personal differences in the first method. The maker's calibration of the colorimeter filters was assumed, though only indirectly checked. Finally, the last nine colors were deliberately chosen for the region of worst check found with the first 23 colors of evenly spaced hue. It is reasonable to suppose that if the spacing were uniform throughout, agreement would be better. Nevertheless, it was found that the checks were rather good.

An estimate of a *reasonable expectancy of check* in the case of hue may be obtained as follows: First, it is commonly found that in work with the spectrum colors, about 150 colors are discriminable, including the purples. Since the Munsell hue circuit has 100 steps, one just discriminable step corresponds to $\frac{2}{3}$ of a Munsell hue step. Second, it is a result of general experience with many thousands of matches that the greater the saturation (chroma), the smaller the hue difference which can be discriminated, hue becoming indeterminate at the neutral point. The colors concerned here had chromas roughly $\frac{1}{4}$ to 1/20 those of the spectrum for which the discrimination difference of $\frac{2}{3}$ hue step was found. Hence it would be conservative to consider 1 to $1\frac{1}{2}$ Munsell hue units as a good check. In respect to chroma, preliminary observations by the authors, confirmed approximately by some investigations by Judd, indicate that a chroma difference of one unit is about equivalent on the average to a hue difference of $1\frac{1}{4}$ units (at chroma 6). Hence, a check to within one chroma unit may be considered conservatively as good.

[4] This work extended over a course of several years, during which time the standard neutral lights in general use underwent changes.

IV. Results and Discussion

The results of the two methods are given in Table I. In this table "Hue Number" is the number of the hue in the hundred-step Munsell scale, numbering the middle red as 5, the middle yellow-red as 15, the middle yellow as 25, and so on, zero being the first step in the cycle following the red-purples. "Average observer deviation" is the mean difference between the hue (or chroma) as determined by each individual observer and the average hue (or chroma) of all. There are also given the amounts of the hue and chroma disagreements of the two methods for each color.

Table I. *Hue and chroma of 32 colors determined by two independent methods.*

	Hue number				*Chroma*			
Serial number	First method	Average observer deviation	Second method	Difference of two methods	First method	Average observer deviation	Second method	Difference of two methods
1	5.8	0.4	4.9	−0.9	7.4	0.3	8.2	+0.8
2	5.0	0.1	4.0	−1.0	11.6	0.3	12.3	+0.7
3	5.2	0.1	4.2	−1.0	10.6	0.3	11.2	+0.6
4	5.7	0.4	4.7	−1.0	8.5	0.0	9.5	+1.0
5	6.7	0.5	5.5	−1.2	6.6	0.3	7.9	+1.3
6	24.9	0.1	23.6	−1.3	3.1	0.2	3.5	+0.4
7	23.9	0.3	21.5	−2.4	5.2	0.2	5.7	+0.5
8	24.4	0.5	22.4	−2.0	5.6	0.3	6.0	+0.4
9	24.4	0.3	22.5	−1.9	7.6	0.3	7.6	0.0
10	24.5	0.4	22.5	−2.0	10.1	0.3	9.5	−0.6
11	44.0	0.6	43.0	−1.0	2.1	0.3	1.8	−0.3
12	43.7	0.4	43.8	−0.1	7.1	0.3	7.0	−0.1
13	43.5	0.6	43.8	+0.3	7.4	0.5	9.5	+2.1
14	41.9	0.5	42.9	+1.0	6.2	0.3	6.6	+0.4
15	42 2	0.7	42.8	+0.6	4.9	0.4	5.5	+0.5
16	69.4	0.5	68.8	−0.5	5.5	0.4	6.1	+0.6
17	67.5	0.9	66.6	−0.9	5.9	0.3	7.4	+1.5
18	65.4	0.4	66.0	+0.5	6.2	0.4	7.9	+1.7
19	65.6	0.3	65.5	−0.1	6.1	0.1	6.9	+0.8
20	84.9	0.1	83.0	−1.9	5.6	0.1	4.6	−1.0
21	85.8	0.5	85.9	+0.1	5.1	0.5	3.9	−1.2
22	85.3	0.5	85.2	−0.1	3.8	0.2	3.4	−0.4
23	86.4	0.7	86.7	+0.3	3.0	0.4	3.1	+0.0
24	17.7	0.2	14.5	−3.2	3.2	0.4	3.4	+0.2
25	14.9	0.1	12.6	−2.3	7.0	0.6	7.3	+0.3
26	15.1	0.1	13.1	−2.0	7.3	0.5	7.2	−0.1
27	15.7	0.3	13.3	−2.4	10.0	0.2	11.4	+1.4
28	16.6	0.8	15.4	−1.2	6.2	0.2	6.9	+0.7
29	34.3	0.2	33.5	−0.8	6.2	0.2	5.6	−0.6
30	35.1	0.1	33.7	−1.4	4.6	0.1	4.3	−0.3
31	34.9	0.1	35.6	+0.7	8.0	0.1	7.7	−0.3
32	35.0	0.0	34.1	−0.9	7.8	0.1	7.3	−0.5

The samples were about equally divided between dark and light colors, running roughly from 3 to 7 value. Among the first 23 colors, the hue check was best for the blues and worst for the yellows; hence the last 9 colors, which are yellowish, were added. The check for the yellow-reds proved worst of all, and it was bad for the green-yellows; but in spite of the inclusion of these results the weighted average disagreement of the hues determined by the two methods was 1.1 hue steps. The weighting was by the number of observers. The average chroma disagreement was worst for the blues and best for the yellows, but not bad in any case; the final weighted average difference was 0.5 chroma unit. The average of the deviations of all observers for all colors from the means for the individual colors for 5 (or 3) observers was 0.4 hue and 0.3 chroma units. Another measure of the uncertainty of the first method is given by the *extreme* ranges of the individual results from observer to observer. The average range of the individual extremes for all colors was 1.16 hue steps and 0.95 chroma steps.

V. Conclusions

It is seen that the disagreement between the direct (first) and indirect (second) methods of determination of the Munsell hue and chroma is about equal to the uncertainty of the direct method. Color specification may be accomplished by means of the quasi-psychological method of colorimetry using the Munsell charts with an average overall uncertainty of the order of one Munsell hue step and one-half a Munsell chroma step. The small size of these differences and the good agreement between the direct visual comparison and the roundabout method including spectrophotometry, colorimetry, and the use of graphs for converting tristimulus to Munsell specification, gives confidence in the latter conversion as well as the method utilizing the Guild colorimeter.

Densitometry and Photographic Printing. Illumination of the Negative and Its Effect upon Density*

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The nature of the photographic image is such that the value of density is dependent upon the optical system in which the image is employed. Light-scatter by the embedded silver particles has been thoroughly discussed in the literature of photography, but, for the most part only the extreme systems of densitometry—"diffuse" and "specular" have been considered. This paper summarizes the previous conclusions and extends the discussion to certain intermediate optical systems of practical importance.

DEVELOPED photographic images are composed of silver particles embedded in gelatin. These particles are not distributed uniformly throughout the matrix, but are gathered in groups which may vary in diameter from a few microns to half a millimeter. Out of the non-homogeneous nature of the photographic deposit arise certain difficulties in the correlation of the results of density measurement with the results obtained in the practical uses of photographic materials. Scattering occurs when light passes through a medium containing particles of a diameter comparable with, or greater than, the radiation wave-length and almost all photographic images are made up of such particles. The value of density or the light-stopping power of the image depends, then, not only upon the amount of silver in the deposit and upon its distribution, but also upon the characteristics of the optical system in which it is employed. It is the purpose of this paper to discuss the manner in which light-scattering by the photographic image influences the effective value of density.

An homogeneous light absorber which is non-selective in its absorption throughout the spectral regions of concern in photometry and photographic printing could, generally speaking, be transferred from the optical system of any densitometer to the optical system of any printer with the assurance that its light-stopping power would be the same in both cases. With the silver image, even in the simplest case, where it is desired to correlate the results of contact printing with a "diffuse density" reading which accounts for all of the transmitted light, the effect of scatter cannot theoretically be neglected. To correlate diffuse density readings with printing results obtained in projection printing systems presents still greater difficulties. The complexity of these problems is evidenced by the voluminous literature on the subject of which a brief summary is given below.

THE LITERATURE OF DENSITOMETRY AND PRINTING

In addition to many theoretical papers on the scattering of light by material particles, there have been a number of practical papers treating specifically of photographic densitometry. Callier,[1] in 1909, was the first to deal quantitatively with the question of scattering as it concerns projection printing, although Burton,[2] in 1887, had pointed out that condenser enlargers resulted in increased print contrast because of scatter. He also mentioned that scatter might be a source of error in Abney's photographic photometer. Later, Hurter and Driffield[3] and Abney[4] indulged in an extended argument—Abney maintaining

* Communication No. 535.

[1] A. Callier, *The Absorption and Scatter of Light by Photographic Negatives, Measured by Means of the Martens Polarization Photometer*, Phot. J. **49** (N.S. **33**), 200 (1909).

[2] W. K. Burton, *A Few Optical Points in Connection with the Englargement*, British J. Photography **34**, 618 (1887).

[3] F. Hurter and V. C. Driffield, *Accuracy of the Grease Spot Photometer*, J. S. C. I. **9**, 725 (1890); *The Sector and Grease Spot Photometers and Their Results*, J. S. C. I. **10**, 20, 98 (1891).

[4] W. de W. Abney, *On the Accuracy of the Grease Spot Photometer for Measuring the Density of Photographic Plates, and a Note on the Sector Photometer*, J. S. C. I. **9**, 722 (1890); *Grease Spot Photometer Measurements*, J. S. C. I. **10**, 18 (1890).

the importance of scatter. Chapman-Jones[5] later supported Abney in his contention.

Callier found as an empirical relation between specular (projection) density and diffuse (contact) density that the diffuse value multiplied by a factor greater than unity, which he designated as "q," equalled the specular density. The accuracy of the linear relationship was later questioned by Bloch and Renwick,[6] who found that both for their own data and those of Callier an exponential relation: $D_d = kD_s^M$, in which D_s is the specular density, D_d the diffuse density, and k and M constants, held with much greater accuracy. The validity of this empirical relation has since been checked by the present author[7] for certain cases.

Silberstein and Tuttle[8] in an analytical treatment of the subject found the two values of density related as follows:†

$$10^{-D_s} = \epsilon \cdot 10^{-D_d} + (1-\epsilon) \cdot 10^{-BD_d},$$

where ϵ is for all practical purposes a constant expressing the fraction of the total scattered light which emerges normally or quasi-normally (in the case of projection printing or densitometry this is the fraction entering the printing lens or measuring element) and B = unity plus the ratio of scattering to absorption coefficients. The smaller the value of ϵ the more nearly will the density relation approach the strict proportionality as stated by Callier. When $\epsilon = 0$ the above relation reduces to $D_s = BD_d$, and when $\epsilon = 1.0$, $D_s = D_d$.

The value of ϵ obviously can be varied within the range from zero to unity by changing the conditions of the measurement, but the value of B is an attribute of the emulsion itself.

The papers just discussed deal principally with the two extremes of photographic densitometry or printing—with cases where the system of negative measurement is either completely specular or completely diffuse. For true specular density, the negative is illuminated by collimated light and measured by the normally transmitted component. For true diffuse density, the negative is either diffusely illuminated or, if it is illuminated in any other manner, the *whole* of the transmitted flux is gathered for measurement. In the latter kind of diffuse density measurement, as exemplified by the instrument in which the material is placed over the opening of an Ulbricht integrating sphere, it makes no difference whether the incident light is collimated or diffuse. In the former kind of diffuse density measuring device, as exemplified by the densitometer in which the sample is placed in contact with a perfectly diffusing material, it makes no difference how large a component of the transmitted light is collected. Except for small differences which are caused by inter-reflection between the sample and the diffusing material in the latter type, the two distinct methods of diffuse density measurement yield identical results.

Conditions which lie between the extremes of the specular and the diffuse are to be found both in densitometers and projection printers. Few practical illumination systems answer the rigid requirements of perfect diffusion or perfect collimation, and no receiver of light transmitted by the image, whether it be the measuring element of a densitometer or the projection lens of a printer, because of its finite size, can strictly limit its collection to the normally transmitted component.

There are not many references in the literature to the manner in which effective density varies between the extreme limits of diffuse and specular with practical optical systems. Tuttle and McFarlane[9] have shown how the effective density of motion picture positive film illuminated by

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[5] Chapman-Jones, *On the Principal Causes of Discrepancies in Opacity Measurements*, Phot. J. **39** (N.S. **23**), 102 (1898).

[6] O. Block and F. F. Renwick, *The Opacity of Diffusing Media*, Phot. J. **56** (N.S. **40**), 49 (1916).

[7] C. Tuttle, *The Relation between Diffuse and Specular Density*, J. Opt. Soc. Am. **12**, 559 (1926).

[8] L. Silberstein and C. Tuttle, *The Relation between the Specular and Diffuse Photographic Densities*, J. Opt. Soc. Am. **14** and Rev. Sci. Inst. 365 (1927).

† It is sometimes more convenient to express this relationship in terms of diffuse and specular transparency, (T_d and T_s) in which case,

$$T_s = \epsilon T_d + (1-\epsilon) T_d^B,$$

where ϵ and B have the same significance as above.

[9] C. Tuttle and J. W. McFarlane, *The Measurement of Density in Variable Density Sound Film*, J. Soc. Mot. Pict. Eng. **15**, 345 (1930).

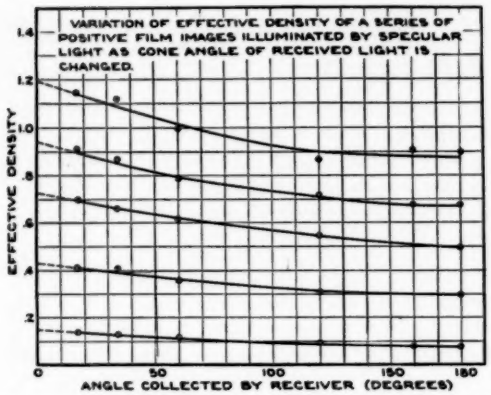

FIG. 1. Density as a function of the angle subtended at the sample by the measuring element.

nearly parallel light varies as the solid angle of the transmitted light collected by the receiver is changed. Effective density as a function of the solid angle of the light collected by the receiving element has been calculated from their data, and is shown graphically in Fig. 1.

From these data, it appears that when the image is illuminated by nearly parallel light, no practical projection printing lens could collect a cone of light of angle wide enough to reduce the effective density appreciably from the true specular value. Assume, for example, the extreme case of a wide-aperture projection printing lens, e.g., one rated at $f/4.0$, working at infinite magnification. The angle subtended by the diameter of such a lens at the sample is about 14 degrees. The effective density with this collected angle is only about three percent less than the true specular density obtained by extrapolation of the curves in Fig. 1 to zero degrees.

The question of the degree of "diffuseness" of the incident light and its effect upon the measured value of density has been considered briefly by Tuttle and Young[10] for the case where only the normal and nearly normal components of transmitted light are collected and measured. Since more complete data of this nature are to form part of the present experimental work, their results will not be discussed at this point.

Although the influence of scatter upon the value of density is obviously of greater importance in projection than in contact printing, there are many references in the literature which lead one to believe that the effect is not negligible in contact printing. At first thought, it would appear that a positive material brought into close contact with the silver image of the negative would collect all the transmitted light as effectively as would the diffuse densitometer of the integrating sphere type. Direction of the scattered light might, however, have some effect upon the positive exposure. If some of the light emerging from the negative strikes the positive at near-grazing incidence, the effectiveness of this light in exposing the positive might be decreased.

Relations between contact printing density (D_ϕ) and visual diffuse density (D_d) have been frequently measured and reported. For the most part, it has been agreed that the two values are linearly related, that is, that $D_\phi = PD_d$ in which P has been named the "printing coefficient."

One would expect in cases where the image is colored by staining developers that P might have some value other than unity but the variety of results quoted below is somewhat surprising in that most of them were obtained for unstained images. Some of the values of the printing coefficient which have been published are: 0.8 (Driffield[11]), 1.0 (Cartwright[12]), 0.92 (Toy[13]), 0.92 and 0.95 (Ferguson[14]), 1.0, 1.14, 1.2, 1.4 (MacKenzie[15]).

None of these observers has attempted to isolate the non-homogeneity effect from the other possible causes of disagreement between printing and measured density values.

DENSITOMETRY OF PRACTICAL PROJECTION PRINTING SYSTEMS

The importance to the practical worker of the relations between optical system characteristics,

[10] C. Tuttle and D. A. Young, *Illumination in Projection Printing and Motion Pictures*, J. Soc. Mot. Pict. Eng. **19**, 842 (1932).

[11] V. S. Driffield, *The Principles Involved in Enlarging*, British J. Photography **41**, 724 (1894).

[12] H. Mills Cartwright, *The Measurement of Photographic Density. I*, Phot. J. **64** (N. S. **48**), 180 (1924).

[13] F. C. Toy, *Standardization of Photographic Density Measurement*, Phot. J. **65** (N.S. **49**), 164 (1925).

[14] W. B. Ferguson, *The Standardization of Measurements of Photographic Density*, Phot. J. **66** (N.S. **50**), 294 (1926).

[15] Donald MacKenzie, *Straight-line and Toe Recording with the Light-Valve*, J. Soc. Mot. Pict. Eng. **17**, 172 (1931).

structure of the photographic image, and final results becomes apparent from a study of the literature. Results of an investigation of the effects of image non-homogeneity in certain representative printing systems should prove of some interest.

Illumination systems for projection printing in actual use range all the way from the highly specular source consisting of a concentrated filament lamp imaged in the projection lens by a condenser to the highly diffuse source achievable through the use of a variety of different optical components.

The degree of diffuseness of negative illumination can be varied throughout a wide range by the simple expedient of introducing beneath the negative, materials which scatter light more or less completely.

Fig. 2 shows a dimensioned sketch of the condenser-relay printer which was assembled for the purpose of obtaining the data which will be reported. The condenser, C, forms a filament image in the projection lens, L, which in turn images the negative, N, in the positive plane, P. At P is located a light-sensitive cell, S, which is calibrated with a high sensitivity galvanometer to read intensity in the printing plane; M is a mask with a hole 1 cm in diameter which is imaged on the cell. The plate, D, immediately beneath the negative is either of clear glass or of diffusing material. By sliding ground or opal glass into the position of D, the diffuseness of the negative illumination may be altered. The diffusing material is placed at a distance below the negative sufficient to prevent its surface being imaged on the printing plane. The cell may be removed from the printing plane and a sheet of sensitive positive material may be substituted for it. It is thus possible to check the results obtained photoelectrically by employing the usual methods of photographic photometry.

It seems of interest to present here density data for two different photographic emulsions, a fine-grained material represented by Positive film and a coarser grained material represented by Supersensitive Panchromatic. Uniformly exposed areas of varying density were prepared from each of these materials. The effective densities of these areas were measured in the projection system of Fig. 1 with various diffusing

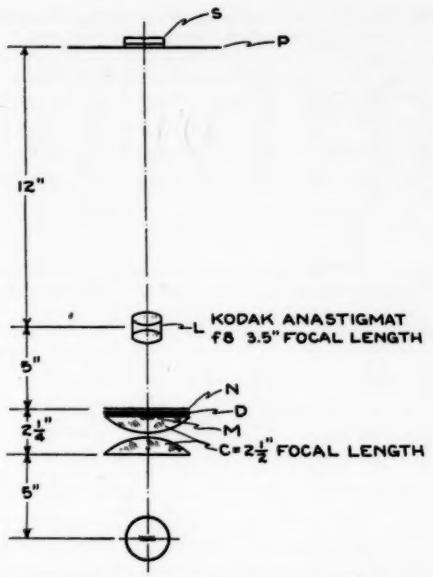

FIG. 2. Projection optical system used in making measurements.

materials beneath the negative. In Tables I and II, the measured values of density are reported in the columns headed "D_1(obs)$\cdots D_5$(obs)." The materials used beneath the negative for the five degrees of diffuseness are as follows: D_1—clear glass plate (no diffusion), D_2—glass plate ground on one side with No. 220 emery powder,* D_3—glass plate 2 mm thick ground on both sides with No. 220 emery, D_4—thinly flashed opal, the optical density of which in a diffuse densitometer was 0.2, D_5—dense pot opal one millimeter thick. The last value, D_5 is a close approximation to true diffuse density.

In Table I, for Positive emulsion, it appears that a simple linear expression represents fairly well the relation between D_5 and the densities effective in the other kinds of illumination.

The expression, $D_e = RD_5$, in which D_e represents any value from D_1 to D_4, and R is a constant chosen for each negative illumination, does not fit the observed data as accurately as the exponential form first published by Bloch and Renwick.[6]

* The number 220 emery powder was selected because it gives the coarsest grinding which will not be imaged in the printing plane as an objectionable non-homogeneity. In general, the coarser the grinding, the more effective will be the diffusion.

TABLE I. *Measured and computed density of Positive film emulsion in various kinds of illumination.*

D_1(obs)	D_1(calc)	D_2(obs)	D_2(calc)	D_3(obs)	D_3(calc)	D_4(obs)	D_4(calc)	D_5(obs)
0.14	0.14	0.12	0.12	0.11	0.11	0.11	0.11	0.09
0.23	0.23	0.22	0.21	0.20	0.19	0.18	0.18	0.15
0.39	0.41	0.38	0.38	0.34	0.35	0.31	0.32	0.28
0.64	0.68	0.61	0.62	0.55	0.56	0.51	0.53	0.46
0.98	0.98	0.92	0.91	0.82	0.82	0.79	0.76	0.68
1.41	1.42	1.31	1.30	1.19	1.17	1.15	1.11	0.99
1.86	1.86	1.72	1.70	1.58	1.53	1.46	1.45	1.30
2.30	2.28	2.10	2.08	1.85	1.88	1.79	1.77	1.60

TABLE II. *Measured and computed density of Supersensitive Panchromatic emulsion in various kinds of illumination.*

D_1(obs)	D_1(calc)	D_2(obs)	D_2(calc)	D_3(obs)	D_3(calc)	D_4(obs)	D_4(calc)	D_5(obs)
0.15	0.16	0.13	0.16	0.11	0.13	0.10	0.12	0.08
0.28	0.28	0.26	0.27	0.22	0.23	0.21	0.21	0.15
0.44	0.44	0.42	0.41	0.36	0.36	0.33	0.33	0.24
0.61	0.63	0.57	0.59	0.50	0.51	0.46	0.47	0.36
0.81	0.81	0.75	0.76	0.68	0.67	0.62	0.61	0.48
1.01	1.02	0.94	0.97	0.85	0.85	0.78	0.74	0.63
1.20	1.22	1.13	1.14	1.00	1.00	0.93	0.93	0.77
1.37	1.35	1.30	1.27	1.13	1.10	1.03	1.00	0.86
1.58	1.57	1.50	1.48	1.30	1.29	1.18	1.17	1.02

For the data of Table II, Supersensitive Panchromatic emulsion, the linear expression is inadequate but the exponential form fits very well.

In Tables I and II, the column headed "D_1(calc)$\cdots D_4$(calc)," have been computed from the relation, $D_e^M = kD_5$. The exponent, M, has a value of 1.02 for Positive emulsion and 1.11 for Supersensitive Panchromatic. The values of k for the different systems of negative illumination are shown in Table III.

TABLE III. *Coefficients in the equation relating diffuse density with density effective in various optical systems.*

Photographic material	1 No diffusion	2 Ground glass	3 Double ground glass	4 Flashed opal
Positive	1.45	1.32	1.19	1.12
Supersensitive Pan.	1.62	1.51	1.30	1.17

SPECIFICATION OF THE DEGREE OF DIFFUSENESS OF NEGATIVE ILLUMINATION

Data such as have been presented in the foregoing section assume a greater practical importance when they are associated with some numerical value specifying the degree of diffuseness of the negative illumination.

Data for the brightness of an illuminated diffuser as a function of the angle of observation can, of course, be obtained with the aid of a goniophotometer. They can be shown graphically as has been done in Fig. 3.

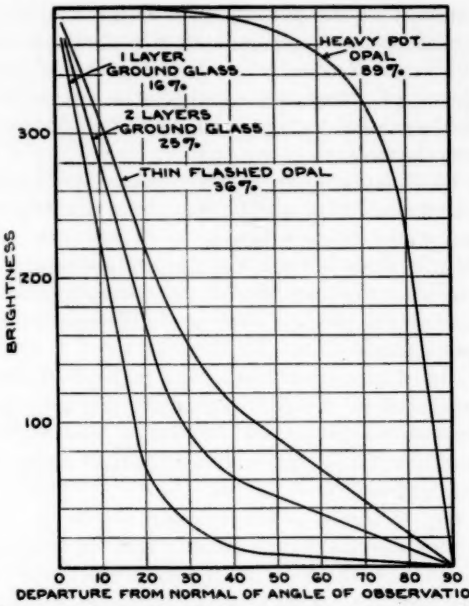

FIG. 3. Brightness distribution about the axis of the optical system with various diffusers.

With distribution which is symmetrical about the normal axis, the data need be obtained for only a single quadrant in a single plane. Since it is brightness distribution and not relative intensity which is of interest, all of the curves have been reduced to the same maximum brightness value. For many practical purposes, a single constant can be used satisfactorily to specify the degree of diffuseness. As previously suggested by Tuttle and Young,[10] a value of diffuseness or, as it may be called, "diffusion efficiency" may be obtained by dividing the area under the brightness distribution curve for the sample into the area under the curve for a perfect diffuser. (The curve for a perfect diffuser is by definition a straight line parallel to the axis of abscissae.) The value of "diffusion efficiency," expressed in percent, is shown on the curves in Fig. 3 for each of the materials used in the previous section.

DENSITOMETRY IN CONTACT PRINTING

It has been shown that the non-homogeneity of photographic images may cause large numerical differences between the results of densitometry

and of projection printing unless the optical systems of measurement and of printing are comparable.

In contact printing, it seems simpler to use similar systems in densitometry and in printing. The usual densitometer light source, if it is designed to diffuse the light fairly well, should certainly give a density reading close to that effective in contact printing.

One question which should be answered is: Can the lack of image homogeneity itself be accountable for discrepancies between diffuse densitometry and contact printing?

Assume two materials, one of which is homogeneous and the other made up of particles which scatter light. Assume that in identical beams of incident light both transmit the same total quantity of actinic light. Will the positive material placed in contact with either of these materials integrate the light transmitted regardless of its direction so that the photographic effect will be the same?

A direct test of this question has been undertaken in the following manner. Two series of densities were made up in the form of a step tablet, one of cut pieces of "neutral gray" gelatin (Wratten filter No. 97) and one of Supersensitive Panchromatic emulsion developed in a non-staining developer. The two tablets were mounted side by side on glass.

Neither of these materials may be assumed to be entirely neutral in color, but both are sensibly so throughout a limited spectral region.

To limit the spectral region used in performing the experiment, a combination of Wratten filters Nos. 47 and 4 was chosen.

Throughout the region isolated by this combination filter, 460 mμ to 520 mμ, it was ascertained by spectrophotometric measurement that both tablet materials were practically non-selective.

Visual densities of both tablets were determined for light passed by the filter combination with a densitometer in which the samples were placed in contact with opal glass. The particular densitometer used was checked against an integrating sphere type of instrument and found to be correct except for very low densities where the inter-reflection between the sample and opal gave rise to readings slightly lower than those obtained with the sphere.

The mounted tablets were illuminated by a tungsten lamp at about twelve inches' distance and contact prints were made on matte, semi-gloss, and glossy developing-out papers. Densities produced on these papers were read on a reflection densitometer and the values were plotted against the diffusely measured tablet densities. A factorial difference in effective densities of the two tablets would, of course, be evidenced by a difference in gradient of the two curves.

No difference in the curves resulting from the gelatin and the photographic tablet prints could be discovered. This was true regardless of the surface characteristics of the positive materials. It appears, therefore, that light-scattering even in the amount appertaining to a coarse-grained material has no measurable influence upon the value of density effective in contact printing.

It should follow from this that the "diffuseness" or specularity of the printing light in contact printing should have no effect on the photographic results. If, however, the diffuse light is produced by means of opal glass in contact with the negative, somewhat surprising results are obtained, as is evidenced by the following experiment.

A photographic negative step tablet was printed by contact, using, first, parallel light, and, second, light diffused by a sheet of flashed opal glass in contact with the negative tablet on the side toward the light source. Precautions were taken to make certain that the light incident upon the negative was identical in quality and intensity in both cases.

It was expected that no difference in the contrast of the prints made from the negative under the two different conditions would be found. Repeated tests showed that there was an actual contrast difference. The prints from the negative when illuminated by opal-diffused light showed in every case a slightly *higher* contrast than that resulting from the specular illumination. Results are given in Table IV for different positive materials and different degrees of posi-

TABLE IV. *Effect of opal glass backing for the negative in contact printing.*

γ (opal in contact)	1.83	2.01	2.04	2.00	2.27
γ (clear glass in contact)	1.70	1.85	1.84	1.80	2.06
Ratio	1.08	1.09	1.11	1.11	1.10

tive development. Line 1 gives the slope of the straight-line portion of the curves (positive density *vs.* negative density) for the prints made with opal glass in contact with the negative. Line 2 gives the results for clear glass in contact with the negative. Line 3 gives the ratio of the two slopes which is approximately constant at 1.1.

A search of the literature discloses one case where data concerning a similar experiment are reported. A communication from the Pathé laboratories[16] states that diffusion of the incident light produced by opal glass in contact with the negatives does *not* affect the print contrast. The data given in the Pathé report do, however, show a contrast difference of about the same magnitude as that found in the foregoing experiment. Presumably this difference, if it was observed, was thought to be due to experimental error since the author, from an analogy with projection printing, would doubtless have expected the opposite effect.

It seems probable that the anomalous effect of higher contrast of the print made with the opal glass-illuminated negative can be accounted for on the basis of inter-reflection between the surfaces involved. This effect, which is negligible with the small reflection coefficient of the clear glass in contact with the negative, may become important when a highly reflecting material, such as opal glass, is concerned.

The effect of the opal reflection is, of course, to reduce the effective value of the density in contact printing below that obtained in the densitometer. The lower negative densities are decreased more than the higher densities and consequently the contrast of the print is increased. The actual amount of the contrast increase depends primarily upon the reflection coefficients of the positive material and the opal, and upon the actual value of the negative tablet densities. A tablet of *high* minimum density will show the effect less than will a tablet of the same density range, but having a *low* minimum density.

Summary

The literature of densitometry has treated almost entirely of the two extreme methods of measurement—diffuse and specular. Practically, photographic images are most frequently used in systems which are neither diffuse nor specular. Data are given for two different types of material measured in intermediate systems and it is shown that results obtainable in any system may be correlated with diffuse density measurements by means of a single formula involving two constants.

Experimental evidence was obtained which showed that a contact printing system is identical with that of a diffuse densitometer and that image non-homogeneity plays no measurable part in the densitometry of contact printing.

[16] Pathé-Cinema Research Laboratories, *Effect of Diffusion of Light in the Density Measurement and in Photographic Printing*, British J. Photography **72**, 474 (1925).

Negative Illumination in Projection Printing*

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(Received August 8, 1934)

Diffusion in the illumination of a negative to be printed by projection affects the image contrast, the print graininess, the delineation of scratches, and the quantity of light available for printing. These four factors are discussed separately and, from the quantitative results obtained, certain general conclusions regarding the required amount of diffusion for a given type of enlarger are drawn.

THE degree of "diffuseness" of the light incident upon a negative to be printed by projection has a marked effect upon several properties of the projected image. As the negative illumination is made more diffuse, the image contrast decreases, the graininess of the print and the images of negative scratches on the print become less apparent and, in general, the illumination intensity available for printing is diminished.

It is of importance to the designer or the purchaser of a projection printer to be able to evaluate these effects with a view to choosing the amount of diffusion best suited to his particular needs.

PRINT IMAGE CONTRAST

The optical density of a non-homogeneous material such as the photographic silver image is dependent upon certain properties of the optical system employed in its measurement, a factor which accounts for the variation of image contrast with diffuseness of illumination. This subject has been treated at some length in a previous paper[1] and at this point it is necessary only to apply certain of the data described there to the problem of projection printing.

Consider a projection printing system such as that shown in Fig. 1, consisting essentially of a light source, a condenser which images the source in the projection lens, the negative to be printed placed close to the condenser, a projection lens, and a positive or printing aperture. If provision is made for inserting between the condenser and negative, a series of diffusing materials, which scatter the light incident upon the negative to greater or lesser extent, a series of prints of varying contrast may be made from any given negative.

Suppose that a negative composed of areas of different density is printed on a bromide paper. A family of reproduction curves like that of Fig. 2 may be obtained. Diffusely measured or contact print densities of the negative determine the abscissa or log E scale, and reflection densities of the print are plotted as the ordinate. The upper curve, marked "1," is the result obtained when the negative step tablet is either printed by contact or by projection with light which passes first through a good diffuser, such as pot opal glass beneath the negative. Curves 2, 3, 4 and 5 result when flashed opal glass, glass ground on both sides, glass ground on one side, and clear glass are used successively between condenser and negative. The curves of the figure are based on data for a fine-grained

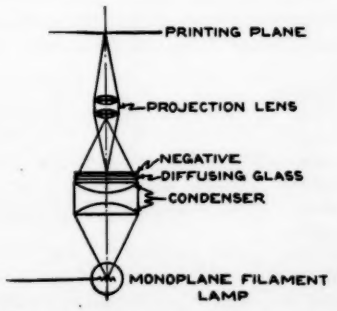

FIG. 1. Projection printer optical system.

* Communication No. 536.

[1] C. Tuttle, *Densitometry and Photographic Printing. Illumination of the Negative and Its Effect upon Density*, preceding paper.

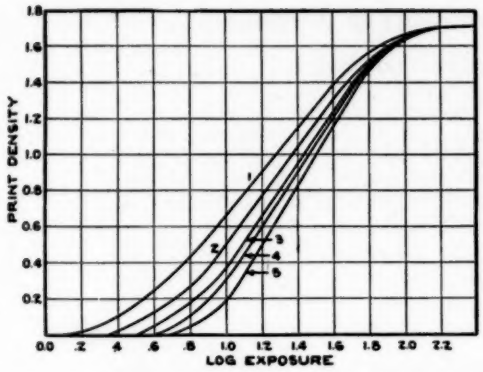

FIG. 2. Reproduction characteristics for "normal" bromide paper printed from a negative illuminated with various degrees of diffuseness.

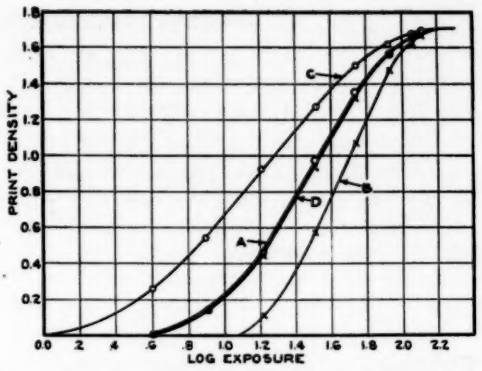

FIG. 3. Reproduction characteristics obtainable on "medium" and "normal" bromide paper printed by diffuse negative illumination (*A* and *C*) and by specular negative illumination (*B* and *D*).

material,* used in making the tablet. Had values for a coarser grained material been assumed, the spread of the curves would have been somewhat greater. These curves show a maximum variation which is equivalent approximately to the step from one contrast grade of bromide paper to the next, i.e., from "normal" to "medium." If prints of a pictorial negative of average contrast were to be made with the two extreme systems, one would have from the pot opal illumination an extremely soft picture and from the specular (condenser) illumination, a fairly hard print.

These data alone are actually not of very great importance in deciding upon the merits of an optical printing system. It is usually possible to produce from a given negative, a projection print of the desired contrast regardless of the printer optical system. This can be done in some printing problems by choosing the contrast grade of the printing paper. The same result can be accomplished by varying the time of print development with certain other types of positive material.

It may be of interest to show, by the use of two examples, that insofar as contrast and tone reproduction are concerned, there is no real difference in prints obtainable with two widely different systems of negative illumination. Assume that the curve marked *A* in Fig. 3 represents the tone reproduction characteristic desired for the print from a certain negative. This curve is the characteristic curve of a "medium" bromide paper and it represents the results obtainable by printing a negative upon this material either by contact or by projection with diffuse negative illumination. If the same negative is specularly printed—with a condenser-relay printer—a projection print of higher contrast such as curve *B* will be obtained. It is possible, however, to choose a paper of inherently lower contrast such as "normal" bromide of which *C* represents the characteristic curve. A specular print of the negative on normal bromide paper gives the curve *D*. This is so close to the desired curve, *A*, that probably no difference in the contrast of the prints would be observed.

The second example, Fig. 4, deals with the same sort of case except that a typical Eastman Positive Emulsion is used as the positive

* The data for Eastman Positive Emulsion as obtained in the previous paper[1] are assumed. The series of effective density values shown below are taken from Table I of that paper.

Density diffusely measured:	0.09	0.15	0.28	0.46	0.68	0.99	1.30	1.60
Density in projection system:								
With flashed opal glass	0.11	0.18	0.32	0.53	0.76	1.11	1.45	1.77
With double ground glass	0.11	0.19	0.35	0.56	0.82	1.17	1.53	1.88
With single ground glass	0.12	0.21	0.38	0.62	0.91	1.30	1.70	2.08
With no diffusion	0.14	0.23	0.41	0.68	0.98	1.42	1.86	2.28

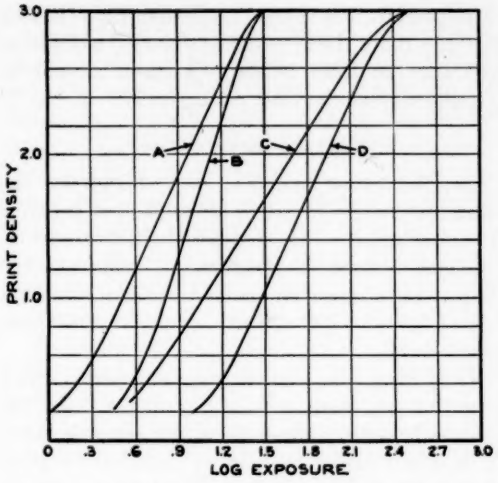

FIG. 4. Reproduction characteristics obtainable on positive film at different development times printed by diffuse negative illumination (*A* and *C*) and by specular negative illumination (*B* and *D*).

material. *A* is the curve obtained when the negative is diffusely printed upon the material and print development is carried to 8 minutes. *B* is the reproduction of the same negative printed by the specular system and developed for the same length of time. *C* is the characteristic curve of the same positive emulsion developed only 3 minutes. *D* is the reproduction obtainable when the negative is semi-specularly printed and the print developed 3 minutes. So far as the contrast is concerned, print *D* almost exactly matches print *A*.

There is an interesting point to be observed in connection with Fig. 4. At the lower time of development, the effective speed of the positive material is considerably decreased. To produce the print represented by *D* requires approximately eight times as much exposure as to produce the print represented by *A*. Obviously then, if the reason for decreasing the diffuseness of illumination is to increase the intensity in the printing plane, some of the benefit is lost because of the lower speed of the material at the lower contrast. It might even happen that prints could be made in the diffuse system in a shorter time than would be possible in the more specular system.

An analogous argument cannot be applied to the printing on different grades of paper but is of interest only when the print contrast is controlled by development time.

PRINTING RANGE

A secondary result which follows from the variation of the effective density with the diffuseness of the negative illumination may in some cases be a matter of considerable importance. The more diffuse the printing system, the smaller will be the ratio of maximum to minimum exposure or, in other words, the exposure range required to print extreme negatives. Negatives of printable quality vary in diffusely measured average density over a range of about 1.6. The densest negative will, therefore, require about forty times the printing exposure of the thinnest negative when prints are made on a diffusely illuminated printer.

To print the same negatives on a specular printer will require that the range be increased from 40 to 1 up to 175 to 1 for fine-grain materials and up to 375 to 1 for coarser grain images. So great an exposure range may be difficult to accomplish or inconvenient to use.

PRINT GRAININESS

It is generally believed that print graininess decreases as the negative illumination is made more and more diffuse. Since it is well known that the appearance of graininess is worse the higher the print contrast, it seemed possible that the increase of contrast caused by specular printing might alone be accountable for objectionable graininess, i.e., that lack of diffusion results in high print contrast and that increased graininess is the result only of increased contrast.

Since the quality of the specular or semi-specular print as specified by the contrast would be the same as that desired for the diffuse illumination print, it seemed desirable to determine the relative graininess of prints made with different degrees of diffuseness of negative illumination but being of the same quality with respect to contrast.

Projection prints on plates were made of a fine-grain negative step tablet. In order to make the graininess of the prints easily measurable, a magnification higher than that ordinarily used in

printing was employed. Various diffusing materials were inserted behind the negative tablet to vary the diffuseness of the illumination. The exposure and development times were so chosen that these prints, step for step, were identical in density.

Relative graininess of each comparable step was then determined in the "graininess apparatus" described by Jones and Deisch,[2] an instrument in which means is provided to increase the viewing distance of the sample until the non-homogeneity is no longer visible. Results of the measurements which are given in Fig. 5 are only relative. No attempt should be made to correlate the numerical values with those published by the originators of the method of measurements.

The abscissa axis of the curve, Fig. 4, is labelled "Diffusion Efficiency," a value determined for each diffusing material as described in the previous paper.[1]

It appears from these data that graininess is continuously improved as the diffusion efficiency is increased. The curve is approximately logarithmic and graininess is approximately halved for every 12 percent increase in diffusion efficiency.

The amount of diffusion which is desirable from the point of view of graininess elimination is obviously a function both of the graininess of the negatives to be dealt with and of the magnification of the image. Inspection of trial prints is perhaps the only reliable criterion of the amount of diffusion needed for a given problem in enlargement printing. Since halftone engravings do not give a true representation of graininess, it is unsatisfactory to reproduce here prints to use as samples and the reader must rely to a large extent upon his own experiments. However, upon the basis of the results obtained in this experiment, it is perhaps permissible to make a general statement as follows: In the case of fine-grain negative material such as Panatomic film, at an enlargement not exceeding four times, graininess would be practically invisible, if the diffusion efficiency of the negative illumination is 40 percent. For coarser grain materials (having perhaps twice the inherent graininess) a diffusion efficiency of from 50 to 60 percent should be satisfactory at the same enlargement.

Scratch Elimination

An important function of diffusion in the illumination of a negative for projection printing is to reduce the contrast with which abrasions on both surfaces of the film are imaged in the print. In a specular print, surface scratches appear as sharply defined black or sometimes white lines.

Obviously the contours of scratches which may be produced upon a negative are myriad and each depth and shape of scratch may have a different effect in each system of negative illumination. The work involved in a comprehensive study of this subject could hardly be justified but a simple experiment serves to illustrate the value of diffusion in an optical system.

A series of scratches was made in the emulsion surface of a step tablet negative. The scratches

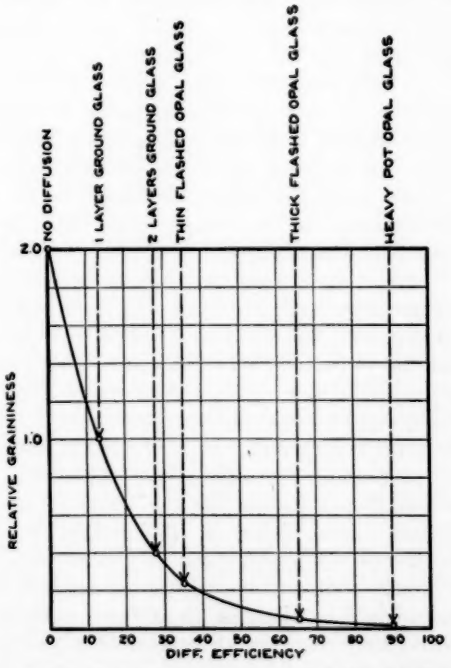

FIG. 5. Effect of diffuseness of negative illumination on print graininess.

[2] L. A. Jones and N. Deisch, *The Measurement of Graininess in Photographic Deposits*, J. Frank. Inst. **190**, 657 (1920); British J. Photography **67**, 689 (1920).

were produced by a pyramidal steel point which was supported by a lever. The depth to which the point entered the emulsion was variable by means of weights which could be attached to the lever. An enlarged print was made of the scratched negative by using highly diffused illumination. One of the lighter scratches was found to be entirely eliminated in the print. The diffuseness of the negative illumination was then varied by steps to the point where it was purely specular. At each step, a print was made and developed to a contrast which matched that of the first print. The scratch which was entirely eliminated in the diffuse print began to appear as the diffusion was decreased. The contrast of the scratch with its background at each step was determined with the aid of a microdensitometer and the data relating diffusion efficiency with the density difference of the scratch and its background are shown in Fig. 6. In the case of the scratch image as in the case of print graininess there appears to be a continuous improvement with increasing diffusion. It is of course impossible by means of these data to specify the necessary amount of diffusion for all printing systems. This value is dependent upon the severity of the abrasions to be expected on the negatives which must be printed. It does appear from these data and from a number of qualitative observations on accidently produced negative scratches that most of the possible improvement occurs before a diffusion efficiency of about 50 percent is reached. In practical printing problems it is probably safe to say that a diffusion efficiency of 50 to 60 percent is sufficient from the point of view of scratch elimination.

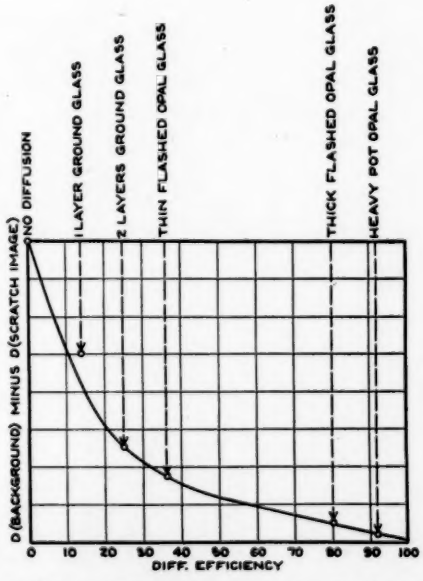

FIG. 6. Effect of diffuseness of negative illumination on the contrast of a scratch image.

INTENSITY OF ILLUMINATION IN THE PRINTING PLANE

Three of the factors just discussed argue strongly for a high degree of diffusion in the illumination of the negative in projection printing. One factor, however, argues against it. This is the sacrifice of light which is entailed by the introduction of diffusing materials. High efficiency of the illumination system is a matter of economy. It saves wattage of the light source, exposure time, or printing material cost.

It is then of interest to include with this discussion some data regarding the illumination available in the printing plane with various systems of negative illumination. In Table II are shown relative values which can be taken as approximately correct for the specific conditions described.

The value of 100 in the table refers to the intensity in the center of the negative plane with the lamp (a concentrated filament, tungsten source, situated at a distance of 5 inches with no intervening optical system). The succeeding four values in the same column refer to the axial intensity in the printing plane with a condenser-relay system with an $f/4.5$ projection lens imaging the negative (condenser plane) at 1–1. The effective "f" value of the lens is $f/9$.

The second column of the table gives the diffusion efficiency. The third column gives the relative printing intensity. The fourth column (which is really the most important) requires some explanation. It is a well established fact that a printer designed to handle a full range of negatives must furnish an exposure sufficient to print through a diffusely measured density at least as high as 2.0. Measured in the various optical systems, a density which has a diffuse value of 2.0 becomes as shown in Table I. Data

TABLE I. *Printing factor for a diffuse density of 2.00 in various optical systems.*

Diffusion efficiency	Approximate effective density	Approximate printing factor
100	2.0	100
89	2.0	100
36	2.1	125
25	2.3	200
16	2.5	300
0	3.0	1000

TABLE II. *Illumination intensity of several printing systems.*

Illumination system	Diffusion efficiency	Intensity in printing plane	Intensity in printing plane with negative density of 2.0
1. Lamp at 5″—contact printing in negative plane	100	100	1.0
2. Condenser-Relay System, no diffusion	0.0	34	0.034
3. Condenser-Relay System with 1 layer 220X ground glass beneath negative	16	1.5	0.005
4. Condenser-Relay System with two layer 220X ground glass beneath negative	25	0.5	0.0025
5. Condenser-Relay System with layer of thinly flashed opal glass beneath negative	36	0.25	0.002
6. Condenser-Relay System with pot opal glass 1 mm in thickness beneath negative	89	0.04	0.0004

for a coarse-grained material have been taken in this case.

The gain in intensity affected by the use of less diffusion is obviously offset to some extent by the requirement of additional light to print through the effectively increased density. The third column shows the printing exposure factor—the antilog of density effective in the system. In column 4 of Table II, the intensities of column 3, i.e., the intensity available in the printing plane with no negative, are divided by the printing factor.

CONCLUSION

In conclusion it can be stated that several matters about any printing problem should be considered before deciding upon the type of negative illumination to be used.

If, for instance, an enlarger is designed to handle only fine-grain negatives of low density upon which there will be few surface abrasions, a condenser-relay printer with no diffusion would be quite satisfactory for low magnifications.

For general work, e.g., the enlarging of amateur negatives, both coarse and fine-grain up to 4 or 5 diameters, a diffusion efficiency of about 50 percent should give satisfactory graininess and scratch elimination.

For the printing of very badly scratched negatives, the highest order of diffusion obtainable seems to promise the best results.

Phosphorescence of Glass Solarized by Soft X-Rays

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(Received June 19, 1934)

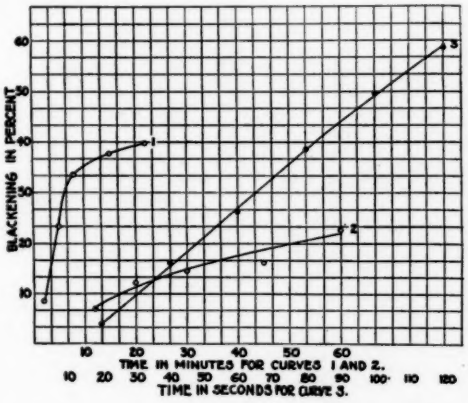
THE method of measuring the intensity of phosphorescence of a substance by exposing a photographic plate to its radiation was used by Le Bon[1] in 1899 for substances which had been caused to phosphoresce by solar radiation, and by Swindells[2] in 1928 for calcium tungstate previously irradiated with x-rays. In making the experiments on the solarization of window glass with soft x-rays,[3] the authors observed the well-known phenomenon of phosphorescence. Since glass may be regarded as a supercooled liquid, and is not crystalline in the same sense as is calcium tungstate, it was deemed interesting to examine the nature of its phosphorescence.

PROCEDURE

The samples of glass,[4] $4.1\times4.1\times0.3$ cm, were placed next to the window of an x-ray tube,[5] at about 3 cm from the focal spot, in the same way as were the samples used for the solarization experiments previously described.[3] The x-ray tube, having a copper target, was operated at 40 kv and 10 m.a. After irradiation the glass samples were placed in a row in close contact with a sheet of Eastman Commercial Ortho Cut Film, solarized side next the emulsion. The film and glasses were kept in a light-tight box, large enough so that the data for one complete experiment could be obtained from the same piece of film. Data for a sensitometry curve were obtained with this same piece. The developed film was cut into as many portions as there were exposures, and each piece run through the Photronic cell apparatus.[3] The percent blackening of the film was taken to be $100-(r/r_0)\times100$, where r is the mean deflection of the galvanometer to right and left when the light came through the black portion of the film, and r_0 that for the clear portion. This blackening is not the same as photographic "density." The experiment consisted of three parts, *viz.*:

(1) Calibration of the film. Curve 3.

(2) Measurement of the variation of the phosphorescence with the time of irradiation of the glass. Five pieces of glass were irradiated for as many different times (from 2 to 22 min.) and placed in contact with the film for 15 hours. The blackening was plotted against time of irradiation (curve 1). This procedure gives what might be called the "phosphorescence yield" of the glass.



Curve 1, blackening of a photographic film as a function of time of irradiation of the glass; curve 2, blackening of a photographic film as a function of time of exposure of film to phosphorescent glow of the glass; curve 3, sensitometry of the film.

(3) Measurement of the variation of the phosphorescence with the time of recovery of the glass (i.e., after cutting off the x-rays). Five pieces of glass were irradiated, each for 15 min., and placed in contact with the film for varying times (from 12 to 60 min.). The blackening was plotted against time of exposure of the film to the glass (curve 2). This procedure gives what might be called the "phosphorescence decay" of the glass.

[1] Le Bon, C. R. **128**, 174–176 (1899).

[2] Swindells, J. O. S. A. and R. S. I. **16**, 165–173 (1928).

[3] Kersten and Dwight, J. Chem. Phys. **1**, 627–629 (1933).

[4] Glass manufactured by the Libbey-Owens-Ford Glass Co. Soda-lime-magnesia window glass, although minor constituents play an important part in solarization effects.

[5] Kersten, Radiology (1934).

In every case one minute elapsed from the time the x-rays were cut off until contact was made between the glass and the film.

Conclusions

(1) The shapes of the "yield" and "decay" curves are the same as those found by Swindells for calcium tungstate when "relative intensity" is plotted against "exposure" (or irradiation), and "time after excitation" (or recovery), respectively.

(2) The essential linearity of the calibration curve permits the ordinates of the yield and decay curves to be considered as being directly proportional to the energy of phosphorescence emitted.

(3) Comparison with the previous investigation[3] shows (a) that curve 1 is similar to that giving the relation between the solarization of the glass and the time of irradiation, i.e., that there is a correlation between solarization and phosphorescence; (b) that curve 2 is similar to that giving the relation between the fading of the solarization and the time after cutting off the x-rays, i.e., that the decay of solarization is associated with the decay of phosphorescence of the solarized part of the glass.

(4) The graphical results given are taken from a single film. Other pieces of the same kind of film give smooth curves of the same general shape but corresponding curves made from different films are not coincident on account of the numerous variables involved.